

Exchange interaction parameters and adiabatic spin-wave spectra of ferromagnets: A “renormalized magnetic force theorem”

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The “magnetic force theorem” is frequently used to compute exchange interaction parameters and adiabatic spin-wave spectra of ferromagnets. The interest of this approach is that it allows to obtain these results from a *non-self-consistent* calculation of the (*single-electron*) *band energy* only, which greatly reduces the computational effort. However, as discussed by various authors, this approach entail some systematic error. Here, a “renormalized magnetic force theorem” allowing to remove this systematic error without increasing significantly the computational effort is presented. For systems with one atom per unit cell, it amounts to a simple renormalization of the spin-wave spectrum. This renormalization greatly improves the agreement between calculated and experimental Curie temperatures of Fe and Ni.

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The *ab initio* study of magnetic interactions in magnetic metals and of interlayer exchange coupling in multilayers has been the subject of a large number of publications during the past few years. Many these studies rely explicitly or implicitly on the use of the “magnetic force theorem” (MFT), which allows to perform the calculations *non-self-consistently*, and by taking into account only *single-particle energies*. This is of great practical interest for it reduces the computational effort by several orders of magnitude.

The force theorem was first introduced for the case of non-magnetic systems [1]. Extensions to the case of magnetic systems have been published by Oswald *et al.* [2], who focused on the case of magnetic impurities embedded in a non-magnetic host, and by Liechtenstein *et al.* [3], who addressed the case of exchange interactions and spin-wave spectra of ferromagnetic systems. However, as mentioned by Liechtenstein *et al.* in their 1984 paper, their MFT yields exact results only in the limit of infinite magnon wavelength. At finite magnon wavelength, their prescription entails some systematic error, as emphasized recently by various authors [4, 5].

In the present Letter I present a “renormalized MFT”, which corrects the systematic error entailed by the MFT of Liechtenstein *et al.* [3] (hereafter called the “bare MFT”). In the case of periodic systems with a single site per unit cell, this results in an extremely simple renormalization of the spin-wave spectrum and Curie temperature. The Curie temperatures calculated with this procedure are found to be in excellent agreement with the experimental ones, both for Fe and Ni.

The problem we want to address here is that of calculating the energy required to create some static transverse deviation of the magnetic moments in a ferromagnet. The physical motivation for this is that, within the adiabatic approximation (i.e., provided that the characteristic time scale for the dynamics of such fluctuations is long as compared with the time scale for the dynamics of

band electrons), it provides a good approximation to the energy of the low-lying magnetic excitations and therefore allows to address from an *ab initio* point of view the thermodynamics of itinerant ferromagnets. In practice this results into a mapping of the complicated itinerant ferromagnetic system onto an effective Heisenberg system having the same low-lying excitations, but much less degrees of freedom. Let $n(\mathbf{r})$ and $\mathbf{m}(\mathbf{r}) \equiv m(\mathbf{r})\mathbf{u}(\mathbf{r})$ be, respectively, the charge- and spin-density. For transition-metal systems, it is usually justified to neglect (as is almost always done) intra-atomic non-collinearity and fluctuations (which are generally expected to cost a high energy). We therefore have $\mathbf{u}(\mathbf{r}) = \mathbf{u}_{\mathbf{R}}$ if \mathbf{r} belongs to the atomic cell $\Omega_{\mathbf{R}}$ around atom \mathbf{R} , and we wish to impose a prescribed direction to the unit vector $\mathbf{u}_{\mathbf{R}}$ of each site \mathbf{R} [6].

The proper way of doing this relies on the “constrained density functional theory” of Dederichs *et al.* [7], which, in the present case, amounts to introduce some local external field $\mathbf{B}_{\mathbf{R}}^{\perp}$, perpendicular to the local magnetic moment axis $\mathbf{u}_{\mathbf{R}}$, playing the role of Lagrange parameters. As we wish to constrain two components of the magnetic moment for each site, we have two Lagrange parameters per site, which are the magnitude and azimuthal angle of the local constraining field.

In (non-relativistic) spin-density functional theory [8], the ground state energy E_0 is obtained by minimizing the Hohenberg-Kohn functional (HKF) $\mathcal{E}_{\text{HK}}[\rho] = \mathcal{T}_0[\rho] + \mathcal{E}_{\text{ext}}[\rho] + \mathcal{E}_H[\rho] + \mathcal{E}_{\text{xc}}[\rho]$, with respect to the spinor-density $\rho \equiv (n\sigma_0 + \mathbf{m} \cdot \boldsymbol{\sigma})$, where σ_0 is the unit spinor and $\boldsymbol{\sigma}$ the vector-spinor whose components are the Pauli matrices. The various terms are respectively, the kinetic energy of a non-interacting system having the same spinor-density, the potential energy, the Hartree part of the Coulomb energy, and the exchange-correlation energy. The constrained ground state energy is found by introducing the new constrained HKF (CHKF) [7] $\mathcal{F}_{\text{HK}}[\rho, \mathbf{B}^{\perp}] \equiv \mathcal{E}_{\text{HK}}[\rho] + \mathcal{E}_{\text{cons}}[\rho, \mathbf{B}^{\perp}]$, where the constraint

term is given by $\mathcal{E}_{\text{cons}}[\rho, \mathbf{B}^\perp] \equiv -\sum_{\mathbf{R}} \mathbf{B}_{\mathbf{R}}^\perp \cdot \int_{\Omega_{\mathbf{R}}} d\mathbf{r} \mathbf{m}(\mathbf{r})$, and minimizing $\mathcal{F}_{\text{HK}}[\rho, \mathbf{B}^\perp]$ with respect to $\rho(\mathbf{r})$, and to the set $\mathbf{B}^\perp \equiv \{\mathbf{B}_{\mathbf{R}}^\perp\}$. As we want to calculate the energy change associated with some *infinitesimal* transverse fluctuation $\delta \mathbf{u}_{\mathbf{R}}$ away from the ferromagnetic configuration, we may attempt to use the variational properties of the CHKF in order to minimize the computational effort. The difficulty lies in the calculation of the kinetic energy term, which requires the knowledge of the effective one-electron spinor-potential $w_{\text{eff}}[\rho] \equiv V_{\text{eff}}\sigma_0 - \mathbf{B}_{\text{eff}} \cdot \boldsymbol{\sigma}$ of energy eigenvalues $\varepsilon_i(w_{\text{eff}}[\rho])$ (labeled in order of increasing energy) and eigenvectors $\psi_i(\mathbf{r})$ which yields ρ as output spinor-density, i.e., $\rho(\mathbf{r}) = \sum_{i=1}^{N_{\text{el}}} \left\{ |\psi_i(\mathbf{r})|^2 \sigma_0 + [\psi_i^\dagger(\mathbf{r}) \boldsymbol{\sigma} \psi_i(\mathbf{r})] \cdot \boldsymbol{\sigma} \right\}$. The kinetic energy is then given by $\mathcal{T}_0[\rho] = \sum_{i=1}^{N_{\text{el}}} \varepsilon_i(w_{\text{eff}}[\rho]) - \int d\mathbf{r} (nV_{\text{eff}} - \mathbf{m} \cdot \mathbf{B}_{\text{eff}})$. This *implicit* dependence of $w_{\text{eff}}[\rho]$ upon the spinor-density ρ is the origin of our difficulties. In fact, starting from a trial *input* potential and magnetic field and solving the one-electron problem, we are only able to obtain the value of the HKF for the *output* spinor-density, which we did not know *a priori*, but not for some spinor-density chosen *a priori*. This problem can be circumvented by using an auxiliary energy functional, first introduced by Harris [9]. The Harris functional (HF) $\mathcal{F}_{\text{Harris}}[\rho, \mathbf{B}^\perp]$, in the present context, has the same form as the CHKF $\mathcal{F}_{\text{HK}}[\rho, \mathbf{B}^\perp]$, with $\mathcal{T}_0[\rho]$ replaced by

$$\mathcal{T}'[\rho, \mathbf{B}^\perp] \equiv \sum_{i=1}^{N_{\text{el}}} \varepsilon_i(w'_{\text{eff}}[\rho, \mathbf{B}^\perp]) - \int d\mathbf{r} (nV'_{\text{eff}} - \mathbf{m} \cdot \mathbf{B}'_{\text{eff}}), \quad (1)$$

where the new effective spinor-potential $w'_{\text{eff}}[\rho, \mathbf{B}^\perp] \equiv V'_{\text{eff}}\sigma_0 - \mathbf{B}'_{\text{eff}} \cdot \boldsymbol{\sigma}$ is *defined explicitly* in terms of ρ and \mathbf{B}^\perp by

$$w'_{\text{eff}}[\rho, \mathbf{B}^\perp](\mathbf{r}) \equiv \frac{\delta(\mathcal{E}_{\text{ext}}[\rho] + \mathcal{E}_H[\rho] + \mathcal{E}_{\text{xc}}[\rho] + \mathcal{E}_{\text{cons}}[\rho, \mathbf{B}^\perp])}{\delta \rho(\mathbf{r})}, \quad (2)$$

where the functional derivative is taken at the *a priori* prescribed spinor density. This essential difference between $w'_{\text{eff}}[\rho, \mathbf{B}^\perp]$ and $w_{\text{eff}}[\rho]$ allows us to calculate *explicitly* the value of the HF for a spinor-density ρ and constrains \mathbf{B}^\perp chosen *a priori* (provided we know some suitable approximation of $\mathcal{E}_{\text{xc}}[\rho]$), which constitutes a great advantage with respect to the CHKF. In view of the Kohn-Sham theorem [8], the HF and CHKF obviously take the same value E_0 for the density ρ^* and constrains $\mathbf{B}^{\perp*}$ corresponding to the constrained ground state (solution of the constrained Kohn-Sham equation). In addition, the HF can be shown to be *stationary* (but not necessarily *minimal*, in contrast to the CHKF) with respect to ρ and \mathbf{B}^\perp in the vicinity of the constrained ground state $(\rho^*, \mathbf{B}^{\perp*})$. The properties of the HF have been studied by various authors who found that, in fact, it often yields a better approximation of the ground state

energy, in the vicinity of $(\rho^*, \mathbf{B}^{\perp*})$, than the corresponding HKF [10].

Starting from the ferromagnetic state (with $\mathbf{u}_{\mathbf{R}}^0 = \mathbf{u}^0$ for all sites \mathbf{R}), for which we assume the self-consistent density $\rho_0 \equiv n_0\sigma_0 + m_0\mathbf{u}_0 \cdot \boldsymbol{\sigma}$ to be known, we perform some infinitesimal rotations: $\mathbf{u}_{\mathbf{R}} = \mathbf{u}^0 + \delta \mathbf{u}_{\mathbf{R}}$. For the ferromagnetic state, the constraining field vanishes everywhere. For the rotated state, we approximate the energy by using the HF $\mathcal{F}_{\text{Harris}}[\rho, \mathbf{B}^\perp]$, evaluated for a trial input density equal to $\rho_{\text{in}} \equiv n_0\sigma_0 + m_0(\mathbf{u}_0 + \delta \mathbf{u}) \cdot \boldsymbol{\sigma}$, and for some trial input constrain $\mathbf{B}_{\text{in}}^\perp$ (to be specified later).

If we use the local density approximation (LDA) for the exchange correlation term (as is almost always done), the only term in our trial evaluation of the HF which varies with $\delta \mathbf{u} \equiv \{\delta \mathbf{u}_{\mathbf{R}}\}$ is the band energy (sum of eigenvalues ε_i), so that the energy associated with the fluctuation $\delta \mathbf{u}$ is

$$\Delta E(\delta \mathbf{u}) = \sum_{i=1}^{N_e} \{ \varepsilon_i(w'_{\text{eff}}[\rho_{\text{in}}, \mathbf{B}_{\text{in}}^\perp]) - \varepsilon_i(w'_{\text{eff}}[\rho_0, 0]) \} + \mathcal{O}_2(\delta n, \delta m, \delta \mathbf{B}^\perp), \quad (3)$$

where $\mathcal{O}_p(x, y, \dots)$ is a quantity of order p (and higher) in x, y, \dots , and $\delta n \equiv n^* - n_0$, $\delta m \equiv m^* - m_0$, $\delta \mathbf{B}^\perp \equiv \mathbf{B}^{\perp*} - \mathbf{B}_{\text{in}}^\perp$, respectively. We are looking for an expansion of $\Delta E(\delta \mathbf{u})$ to second order in $\delta \mathbf{u}$ of the form

$$\Delta E(\delta \mathbf{u}) = \sum_{\mathbf{R}, \mathbf{R}'} A_{\mathbf{R}\mathbf{R}'} \delta \mathbf{u}_{\mathbf{R}} \cdot \delta \mathbf{u}_{\mathbf{R}'} + \mathcal{O}_4(\delta \mathbf{u}), \quad (4)$$

with $A_{\mathbf{R}\mathbf{R}'} \equiv -J_{\mathbf{R}\mathbf{R}'} + \delta_{\mathbf{R}\mathbf{R}'} (\sum_{\mathbf{R}''} J_{\mathbf{R}\mathbf{R}''})$, which *defines* the coupling parameters $J_{\mathbf{R}\mathbf{R}'}$. The sum rule

$$\sum_{\mathbf{R}} A_{\mathbf{R}\mathbf{R}'} = \sum_{\mathbf{R}'} A_{\mathbf{R}\mathbf{R}'} = 0 \quad (5)$$

expresses the fact that the total energy remains invariant (within the non-relativistic theory) upon a uniform rotation of the magnetization. The definition (4) for $A_{\mathbf{R}\mathbf{R}'}$ implies that it is related to the (static) transverse susceptibility χ by: $2A_{\mathbf{R}\mathbf{R}'} = M_{\mathbf{R}} (\chi^{-1})_{\mathbf{R}\mathbf{R}'} M_{\mathbf{R}'}$.

The *bare* MFT of Liechtenstein *et al.* [3] amounts to make the choice $\mathbf{B}_{\text{in}}^\perp \equiv 0$ for the trial input constraining fields. They then obtain

$$\Delta E(\delta \mathbf{u}) = \sum_{\mathbf{R}, \mathbf{R}'} \tilde{A}_{\mathbf{R}\mathbf{R}'} \delta \mathbf{u}_{\mathbf{R}} \cdot \delta \mathbf{u}_{\mathbf{R}'} + \mathcal{O}_2(\delta n, \delta m, \mathbf{B}^{\perp*}), \quad (6)$$

with $\tilde{A}_{\mathbf{R}\mathbf{R}'} \equiv -\tilde{J}_{\mathbf{R}\mathbf{R}'} + \delta_{\mathbf{R}\mathbf{R}'} (\sum_{\mathbf{R}''} \tilde{J}_{\mathbf{R}\mathbf{R}''})$, and

$$\begin{aligned} \tilde{J}_{\mathbf{R}\mathbf{R}'} &\equiv \frac{1}{\pi} \text{Im} \int_{-\infty}^{\varepsilon_F} d\varepsilon \int_{\Omega_{\mathbf{R}}} d\mathbf{r} \int_{\Omega_{\mathbf{R}'}} d\mathbf{r}' B_{\text{xc}}(\mathbf{r}) G^\dagger(\mathbf{r}, \mathbf{r}') \\ &\quad \times B_{\text{xc}}(\mathbf{r}') G^\dagger(\mathbf{r}', \mathbf{r}). \end{aligned} \quad (7)$$

Since δn and δm are even with respect to $\delta \mathbf{u}$, they are generally of second order in $\delta \mathbf{u}$; however, the constrains

$\mathbf{B}^{\perp\star}$ are odd with respect to $\delta\mathbf{u}$, and therefore generally of first order in $\delta\mathbf{u}$, so that $\mathcal{O}_2(\delta n, \delta m, \mathbf{B}^{\perp\star}) = \mathcal{O}_2(\delta\mathbf{u})$, although $\mathcal{O}_2(\delta n, \delta m) = \mathcal{O}_4(\delta\mathbf{u})$. As a consequence, the parameters $J_{\mathbf{R}\mathbf{R}'}$ (hereafter called the *bare* exchange parameters) are *not* equal to the *true* exchange parameters $J_{\mathbf{R}\mathbf{R}'}$ and entail some systematic error. Note, however, that this problem does not occur for the case considered

by Oswald *et al.* [2] since the constraints vanish in the case they consider.

Clearly, to calculate correctly the *true* exchange parameters $J_{\mathbf{R}\mathbf{R}'}$, we need to take the *exact* constraints $\mathbf{B}^{\perp\star}$ (which are still unknown yet) as trial input in our estimate of the HF. If we do so, we obtain after some algebra,

$$\Delta E(\delta\mathbf{u}) = \sum_{\mathbf{R}, \mathbf{R}'} \left[\tilde{A}_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}} \cdot \delta\mathbf{u}_{\mathbf{R}'} + (M_{\mathbf{R}} \delta\mathbf{R}\mathbf{R}' - \tilde{K}_{\mathbf{R}\mathbf{R}'}) \mathbf{B}_{\mathbf{R}}^{\perp\star} \cdot \delta\mathbf{u}_{\mathbf{R}'} - \frac{1}{2} \tilde{\chi}_{\mathbf{R}\mathbf{R}'} \mathbf{B}_{\mathbf{R}}^{\perp\star} \cdot \mathbf{B}_{\mathbf{R}'}^{\perp\star} \right] + \mathcal{O}_4(\delta\mathbf{u}), \quad (8)$$

with $M_{\mathbf{R}} \equiv \int_{\Omega_{\mathbf{R}}} d\mathbf{r} m(\mathbf{r})$ is the magnetic moment of atom \mathbf{R} , and where the *bare* transverse susceptibility $\tilde{\chi}_{\mathbf{R}\mathbf{R}'}$ and the exchange-correlation response function $\tilde{K}_{\mathbf{R}\mathbf{R}'}$ are given by

$$\tilde{\chi}_{\mathbf{R}\mathbf{R}'} \equiv \frac{2}{\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \int_{\Omega_{\mathbf{R}}} d\mathbf{r} \int_{\Omega_{\mathbf{R}'}} d\mathbf{r}' \operatorname{Im} [G^{\uparrow}(\mathbf{r}, \mathbf{r}') G^{\downarrow}(\mathbf{r}', \mathbf{r})], \quad (9)$$

$$\tilde{K}_{\mathbf{R}\mathbf{R}'} \equiv \frac{1}{\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \int_{\Omega_{\mathbf{R}}} d\mathbf{r} \int_{\Omega_{\mathbf{R}'}} d\mathbf{r}' \operatorname{Im} [G^{\uparrow}(\mathbf{r}, \mathbf{r}') B_{\text{xc}}(\mathbf{r}') G^{\downarrow}(\mathbf{r}', \mathbf{r}) + G^{\downarrow}(\mathbf{r}, \mathbf{r}') B_{\text{xc}}(\mathbf{r}') G^{\uparrow}(\mathbf{r}', \mathbf{r})]. \quad (10)$$

The constraints are obtained by expressing the transverse moments $M_{\mathbf{R}} \delta\mathbf{u}_{\mathbf{R}}$ as resulting from the transverse exchange-correlation and constraining fields: $M_{\mathbf{R}} \delta\mathbf{u}_{\mathbf{R}} = \sum_{\mathbf{R}'} (\tilde{K}_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'} + \tilde{\chi}_{\mathbf{R}\mathbf{R}'} \mathbf{B}_{\mathbf{R}'}^{\perp\star})$. In order to keep the expressions compact we introduce matrix notations: \mathbf{A} , $\tilde{\mathbf{A}}$, $\tilde{\mathbf{K}}$, $\tilde{\mathbf{X}}$, $\tilde{\mathbf{M}}$ are the matrices whose $(\mathbf{R}\mathbf{R}')$ -elements are, respectively, $A_{\mathbf{R}\mathbf{R}'}$, $\tilde{A}_{\mathbf{R}\mathbf{R}'}$, $\tilde{K}_{\mathbf{R}\mathbf{R}'}$, $\tilde{\chi}_{\mathbf{R}\mathbf{R}'}$, $M_{\mathbf{R}} \delta\mathbf{R}\mathbf{R}'$. Inserting the resulting expression of the constraints, $\mathbf{B}_{\mathbf{R}}^{\perp\star} = \sum_{\mathbf{R}'} [\mathbf{X}^{-1}(\mathbf{M} - \tilde{\mathbf{K}})]_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'}$, into the above expression of $\Delta E(\delta\mathbf{u})$, we finally obtain the *exact* explicit expression of the *renormalized* exchange parameters:

$$\mathbf{A} = \tilde{\mathbf{A}} + \frac{1}{2}(\mathbf{M} - \tilde{\mathbf{K}}^T) \tilde{\mathbf{X}}^{-1}(\mathbf{M} - \tilde{\mathbf{K}}). \quad (11)$$

One can easily prove the sum rule $M_{\mathbf{R}} = \sum_{\mathbf{R}'} \tilde{K}_{\mathbf{R}\mathbf{R}'}$, which implies that the constraints $\mathbf{B}_{\mathbf{R}}^{\perp\star}$ vanish for a coherent rotation of all magnetic moment, and that the sum rule (5) is indeed satisfied. The above result, Eq. (11), the “*renormalized* magnetic force theorem”, constitutes the main result of this Letter. Its importance is that it corrects the systematic error introduced by the “*bare* magnetic force theorem”, without increasing significantly the computational effort. It therefore provides a general method to calculate *exactly* the exchange parameters $J_{\mathbf{R}\mathbf{R}'}$ which is several orders of magnitude faster than a fully self-consistent calculation.

The linearized equation of motion of the transverse fluctuations is

$$M_{\mathbf{R}} \frac{d\delta\mathbf{u}_{\mathbf{R}}}{dt} = 2 \frac{\partial \Delta E}{\partial \delta\mathbf{u}_{\mathbf{R}}} \times \mathbf{u}^0 = 4 \sum_{\mathbf{R}'} A_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'} \times \mathbf{u}^0, \quad (12)$$

and the spin-wave energies are given, as usual, by the eigenvalues of the symmetric matrix $\hbar\Omega \equiv 4 \mathbf{M}^{-1/2} \mathbf{A} \mathbf{M}^{-1/2}$.

Some better physical insight into the nature of the above renormalization of the exchange parameters can be obtained if one performs a simple, but yet quite reasonable, approximation. Let us define

$$\Delta_{\mathbf{R}} \equiv \frac{2}{M_{\mathbf{R}}} \int_{\Omega_{\mathbf{R}}} d\mathbf{r} B_{\text{xc}}(\mathbf{r}) m(\mathbf{r}) = \frac{4}{M_{\mathbf{R}}} \sum_{\mathbf{R}'} \tilde{J}_{\mathbf{R}\mathbf{R}'}, \quad (13)$$

which can be seen as some average of the exchange splitting on site \mathbf{R} . The second equality, in the above equation expresses a sum rule related to the invariance with respect to a global spin rotation. If the magnetization, within an atomic cell, is sufficiently rigid, i.e., if intra-atomic fluctuations of the spin-density have a large energy cost (as is usually the case in transition metals), one has approximately:

$$\tilde{K}_{\mathbf{R}\mathbf{R}'} \approx \frac{4\tilde{J}_{\mathbf{R}\mathbf{R}'}}{\Delta_{\mathbf{R}}}, \text{ and } \tilde{\chi}_{\mathbf{R}\mathbf{R}'} \approx \frac{8\tilde{J}_{\mathbf{R}\mathbf{R}'}}{\Delta_{\mathbf{R}}\Delta_{\mathbf{R}'}}. \quad (14)$$

Note that the above relations become exact if do not perform the discretization approximation $\mathbf{u}(\mathbf{r}) \rightarrow \mathbf{u}_{\mathbf{R}}$ (see footnote [6]). One then obtains the following simple and transparent renormalization for the exchange interaction parameters,

$$\mathbf{A} = \tilde{\mathbf{A}} \left(1 - 4 \mathbf{M}^{-1} \Delta^{-1} \tilde{\mathbf{A}} \right)^{-1}, \quad (15)$$

and for the spin-wave matrix

$$\hbar\Omega = \hbar\tilde{\Omega} \left(1 - \Delta^{-1} \hbar\tilde{\Omega} \right)^{-1}, \quad (16)$$

where $(\Delta)_{\mathbf{R}\mathbf{R}'} \equiv \Delta_{\mathbf{R}}\delta_{\mathbf{R}\mathbf{R}'}$ and $\hbar\tilde{\Omega} \equiv 4M^{-1/2}\tilde{A}M^{-1/2}$ is the *bare* (unrenormalized) spin-wave matrix. Noting that the bare parameters $\tilde{A}_{\mathbf{R}\mathbf{R}'}$ can be expressed in terms of the Stoner parameters $(I^{\text{xc}})_{\mathbf{R}\mathbf{R}'} \equiv \delta_{\mathbf{R}\mathbf{R}'}\Delta_{\mathbf{R}}/(2M_{\mathbf{R}})$ as $2\tilde{A} = MI^{\text{xc}}(1 - \tilde{X}I^{\text{xc}})M$, one easily shows that the renormalization (15) can be re-expressed as

$$2A \equiv MX^{-1}M = M(\tilde{X}^{-1} - I^{\text{xc}})M. \quad (17)$$

The above expression has the familiar form of the random-phase approximation (RPA) result for the transverse susceptibility; it is important to realize, however, that in the present context (as shown by the above derivation) this result is *exact* within the LDA (except for the discretization approximation $\mathbf{u}(\mathbf{r}) \rightarrow \mathbf{u}_{\mathbf{R}}$, which as already indicated, can be removed easily). The present approach is therefore formally equivalent to approaches based on calculations of the transverse susceptibility [5], however without the need for self-consistent total energy calculations.

For the particular case of periodic lattices, the above equations are most conveniently solved in Fourier space. For systems with a single atom per unit cell, and using the approximation (14), the renormalization of the exchange parameters leads to the simple rescaling of the spin-wave spectrum

$$\hbar\omega(\mathbf{q}) = \frac{\hbar\tilde{\omega}(\mathbf{q})}{1 - \hbar\tilde{\omega}(\mathbf{q})/\Delta}. \quad (18)$$

The above result clearly shows that the *bare* MFT yields correct results only in the limit of long wavelength (in particular, it yields the correct spin-wave stiffness D), or if the spin-wave energies remain much smaller than the exchange splitting Δ . The above result is in agreement with the estimate of the error entailed by the *bare* MFT proposed by Grotheer [5]: $\hbar\tilde{\omega} \leq \hbar\omega \leq \hbar\tilde{\omega}/(1 - \hbar\tilde{\omega}/\Delta)^2$.

The Curie temperature can be calculated by means of the random-phase-approximation (RPA) Green's function method [11]. For periodic systems with a single atom per unit cell, one has

$$\frac{1}{k_B T_C^{\text{RPA}}} = \frac{6}{M} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\hbar\omega(\mathbf{q})}, \quad (19)$$

so that, by using approximation (14), one therefore obtains an extremely simple renormalization of the (RPA) Curie temperature:

$$k_B T_C^{\text{RPA}} = k_B \tilde{T}_C^{\text{RPA}} \left(1 - 6 \frac{k_B \tilde{T}_C^{\text{RPA}}}{M\Delta} \right)^{-1}, \quad (20)$$

where \tilde{T}_C^{RPA} is the Curie temperature obtained from the *bare* exchange parameters. As seen from Table I, the renormalization of the exchange parameters considerably

improves the agreement between theoretical and experimental Curie temperatures of Fe and Ni.

The method discussed here provides a convenient and accurate approach to study the exchange interactions, spin-wave spectra and Curie temperature of complex systems such as disordered alloys, ultrathin films, nanostructures, dilute magnetic semiconductors (e.g., $\text{Ga}_{1-x}\text{Mn}_x\text{As}$), etc.

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- [1] A.R. Mackintosh and O.K. Andersen, in *Electrons at the Fermi Surface*, edited by M. Springford (Cambridge University Press, Cambridge, England, 1980), p. 149; V. Heine, *Solid State Physics* **35**, 1 (1980); M. Methfessel and J. Kubler, *J. Phys.F: Met. Phys.* **12**, 141 (1982); M. Weinert *et al.*, *Phys. Rev. B* **32**, 2115 (1985).
- [2] A. Oswald *et al.*, *J. Phys. F: Met. Phys.* **15**, 193 (1985).
- [3] A.I. Liechtenstein *et al.*, *J. Phys. F: Met. Phys.* **14**, L125 (1984); *J. Magn. Magn. Mater.* **67**, 65 (1987).
- [4] D.M. Bylander *et al.*, *Phys. Rev. B* **61**, 11875 (2000); H. Eschrig, private communication (2002).
- [5] O. Grotheer *et al.*, *Phys. Rev. B* **63**, 100401 (2001); O. Grotheer, PhD Thesis, University of Stuttgart (2002).
- [6] Note however, that such a discretization is by no means necessary and all results given here are straightforwardly extended to continuous variables $\mathbf{u}(\mathbf{r})$, by omitting integrations over atomic cells $\Omega_{\mathbf{R}}$ everywhere, and replacing discrete summations over \mathbf{R} by continuous integrations over \mathbf{r} .
- [7] P.H. Dederichs *et al.*, *Phys. Rev. Lett.* **53**, 2512 (1984).
- [8] P. Hohenberg and W. Kohn, *Phys. Rev.* **136**, B864 (1964); W. Kohn and L.J. Sham, *Phys. Rev.* **140**, A1133 (1965); U. von Barth und L. Hedin, *J. Phys. C: Solid State Phys.* **5**, 1629 (1972).
- [9] J. Harris, *Phys. Rev. B* **31**, 1770 (1985).
- [10] A.J. Read and R.J. Needs, *J. Phys.: Condens Matter* **1**, 7565 (1989); M.W. Finnis, *J. Phys.: Condens. Matter* **2** 331, (1990); I.J. Robertson and B. Farid, *Phys. Rev. Lett.* **66**, 3265 (1991).
- [11] M. Pajda *et al.*, *Phys. Rev. B* **64**, 174402 (2001).

TABLE I: Curie temperature calculated within the RPA by using the *bare* (\tilde{T}_C^{RPA}) and *renormalized* (T_C^{RPA}) exchange parameters, as compared with the experimental value (T_C^{exp}). The *bare* Curie temperature (\tilde{T}_C^{RPA}) is taken from Ref. [11]; the *renormalized* Curie temperature (T_C^{RPA}) is obtained from Eq. (20).

system	\tilde{T}_C^{RPA} (K)	T_C^{RPA} (K)	T_C^{exp} (K)
Fe (bcc)	950	1057	1045
Ni (fcc)	350	634	621 – 631